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Real-time measurement of soot from low-emission diesel engines by laser-induced incandescence

Real-time measurement of soot from low-emission Diesel engines by laser-induced incandescence (LII)

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Abstract

Appropriate diagnostic tools for the characterisation of soot emission from state-of-the-art and future engines must meet a number of challenging requirements, which are discussed in the present paper and which include the possibility for a sensitive in situ and real-time application, the provision of several relevant particulate characteristics, and the ease of application to various test environments.

It is shown that the LI²SA (Laser-Induced Incandescence Soot Analyzer) sensor system ideally fulfils these demands. Some essential features of the system are a data acquisition rate of typically 4 Hz, which makes it suitable for highly-transient processes, a standard concentration range from well below 0.1 mg/m³ up to 200 mg/m³, and the simultaneous determination of specific particle surface. Several examples demonstrate the broad applicability of the system for engine development, qualification of engines in stationary and transient tests, and vehicle exhaust control on chassis dynamometers.

Introduction

Continuously decreasing particulate emission levels from diesel fueled engines during the last decades and the need for a more adequate characterisation of these particles regarding human health effects have lead current standard test methods to their limits. Several deficiencies of gravimetric sampling and other methods concerning sensitivity, temporal resolution, the need for exhaust gas dilution or conditioning, and the lacking possibility to distinguish between different size and chemical fractions demonstrate the need to establish new techniques which are suitable to comply with the demands of future regulations.

One particularly important aspect is a separate consideration of different types of particles with different origin, which roughly can be divided in an soluble and nonsoluble fraction. From the toxicological point of view, this distinction is mandatory because of different biological effect mechanisms. This aspect is particularly important as the application of different types of exhaust treatment systems, which in future will be broadly distributed, results in a strongly varying particle composition for different types of combustion engines.

Secondly, for the solid particle fraction, which is mainly determined by elementary carbon (EC), several studies indicate that the particle mass alone is not appropriate to establish a more effect related regulation. Currently, there is a very controversial discussion on the question which additional parameter(s) should be used for a more comprehensive characterisation [1, 2]. Possible measures are the diffusion equivalent diameter, which is most relevant for lung deposition, the total number of agglomerates in a given size range, and the particle surface area [3], which is most important as a carrier of condensed hydrocarbons and for in-vivo chemistry.

The main requirements (mandatory and desirable) on a modern soot particle measurement technique are in summary:

- EC as the most relevant component (adaptation of emission to air quality standards)
- directly mass related information and additional, effect related quantity (e.g., particle size, particle number, particle surface, ...)
- high sensitivity, applicable for ultra-low-emission engines within all relevant test cycles
- high temporal resolution, investigation of highly transient behaviour, better than about 1 Hz
- highly specific on elemental carbon, no interference with other exhaust components
- no influence of measurement conditions (temperature, dilution ratio), no condensation artefacts, cf., e.g., Ref. 4
- in situ, online data evaluation (applicable also as a development tool)
- easy adaptation to different engines, no dilution tunnel necessary
- easy handling

A favourable technique which meets all these requirements is given by laser-induced incandescence (LII).

Measurement Principle

Basic principle of laser-induced incandescence [5-8] is to heat up the exhaust particles by means of a highly energetic laser pulse to temperatures well above about 4000 K and to subsequently detect the strongly enhanced thermal radiation by an appropriate optical detector. By this, a highly specific detection of the EC fraction is feasible. An interference with other exhaust components can be excluded, as all volatile components are vaporised prior to significant radiation. From the maximum value of the detector signal which is taken in the visible wavelength range the mass concentration of the carbonaceous particle fraction can be evaluated. Additionally, from the incandescence decay which for late detection times is dominated by conductive cooling the specific surface area, or primary particle size (which for the chainlike, fractal structure of typical soot particles emitted from combustion engines is equivalent), of the particles can be determined.

Sensor Concept

The LII measurement technique has been incorporated into a compact sensor, which has been designed and built according to the above mentioned requirements on modern particulate measurement and is commercially available as LI²SA (Laser-Induced Incandescence Soot Analyzer) through ESYTEC GmbH. Core elements of this sensor are a transportable 19" rack comprising a compact and robust Nd:YAG laser @532 nm including power supply and elements for data acquisition and storage, a light guiding arm which transmits the laser light to the sensor head, which consists of a cooled and air-purged metal ring that can easily be mounted to the exhaust pipe, a fast and compact photodetector including all optical components, and a thermocouple to measure the exhaust gas temperature. Thus, the sensor allows real in-situ measurements without the need for any potentially problematic sampling procedures and through the integration of all sensor components offers easy handling without further adjustment of components. The sensor head is schematically shown in Fig. 1: the excitation laser with a pulse duration of a few nanoseconds illuminates the measurement volume perpendicularly to the exhaust gas flow. With present settings the cylindrical investigation volume has a length of about 25 mm and a diameter of 2.5 mm.

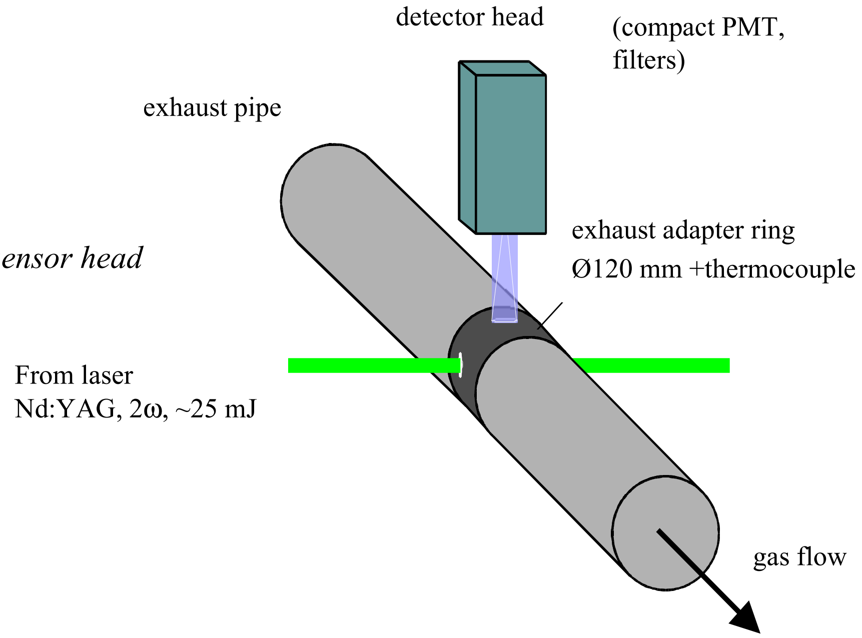

Fig.. 1: Scheme of the sensor head

Another important feature of the present sensor is the capacity for real-time measurements. The temporal resolution of the system is ultimately determined by the repetition rate of the laser, which for the type used is 20 Hz. Although issues of data acquisition and handling and the fundamental advantage that averaging over typically 5 to 20 pulses improves the signal-to-noise ratio and diminishes influences of cycle-to-cycle fluctuations presently suggest data acquisition rates of the order of 1 Hz a simultaneous determination of mass concentration and specific surface (primary particle size) is feasible for the whole standard working range of currently about 0.1-200 mg/m³ on a single-shot basis.

Some Results

The LI²SA sensor described has been employed in numerous applications for particulate measurements in the exhaust gases of a variety of standard and most advanced Diesel engines. The following examples illustrate the capacity of the LI²SA sensor as a useful development tool and a reliable diagnostic system for both engine test benches and chassis dynamometers.

Influence of Rail and Boost Pressure on Particulate Characteristics

A particularly important aspect of the development of modern Diesel engines is the optimisation of injection and exhaust gas recirculation parameters to minimise particulate emission. One special issue is if high pressure injection systems - in spite of a reduction of totally emitted soot mass - are prone to produce potentially hazardous higher number concentrations of soot particles due to reduced sizes. In this context, a production medium-duty truck engine (MAN D0826 L, 6.9l, 206kW) with a common-rail injection system was operated on a standard test bench with various injection pressures at constant speed and load. Two different boost pressures were realised by adjustable turbine wheels. The rail pressure was varied from 60 MPa up to 140 MPa, keeping the injection fuel quantity and the start of injection constant, yet allowing longer injection times for decreased rail pressures.

As expected, higher rail pressures lead to a decrease of the soot mass concentration, due to better spray atomisation and a more homogeneous mixture formation. Higher boost pressures also result in lower concentrations, as higher turbulence rates are induced within the air inlet pipe and improve mixture formation. Additionally, also smaller particles are expected for higher injection pressures (cf. Fig. 2). Yet, from a combination of these results, it can be seen that the decrease in total mass overcompensates for the reduction in particle size resulting in a decrease of the total number concentration with increasing rail and boost pressures.

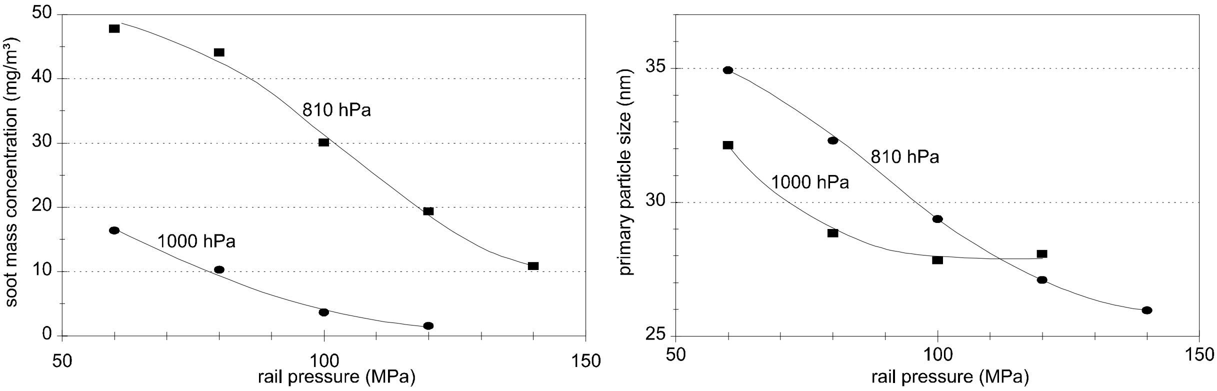

Fig. 2:. Soot mass concentration and primary particle size of a truck engine at two different boost pressures as a function of rail pressure.

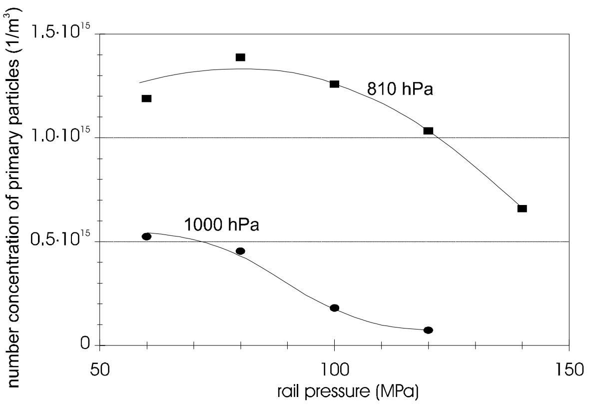

Fig. 3:. Number concentration of primary particles at two different boost pressures as a function of rail pressure

Stationary Cycle Tests

For emission certification, truck engines in Europe must fulfil a 13-mode, steady state procedure, called European Stationary Cycle (ESC) test, comprising a series of defined operating points at various speeds and loads and including a point at low idle operation. The LI²SA system has been employed to characterise the behaviour of the above mentioned truck engine during this test cycle providing information on soot mass concentration (which is related to the Filter Smoke Number, FSN) and additionally on primary particle size. The results of these investigations are depicted in Fig. 4, which indicate that total emission is well below 15 mg/m³ corresponding to 0.85 (FSN) for all operation points and typical primary particle sizes for non-idle conditions are in a range of about 25 - 30 nm.

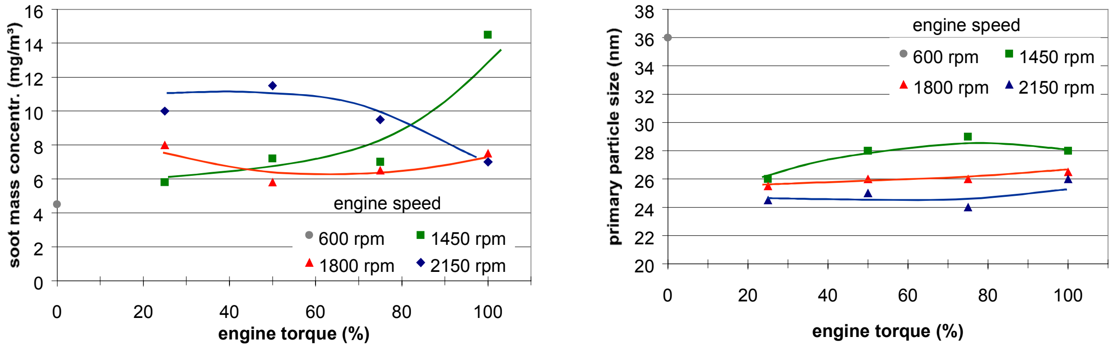

Fig. 4:. Soot mass concentration and primary particle size of soot particles emitted by a truck engine during ESC tests.

It should be noted that the data provided by the LI²SA system provide good correlation with standard measurements on particulate measurements [9]. As an example for a passenger car, results for a modern direct-injection diesel engine (BMW 2.0l, 100kW) are shown in Fig. 5, where LII measurements are compared with a standard smoke meter (AVL 415S). Measurements were performed at (quasi-)

stationary conditions for an operation range from 800 to 4000 1/min and 50 to 250 Nm, respectively, with sampling rates of 1200 single measurement points per minute for the LI²SA system and two measurements per minute for the AVL system.

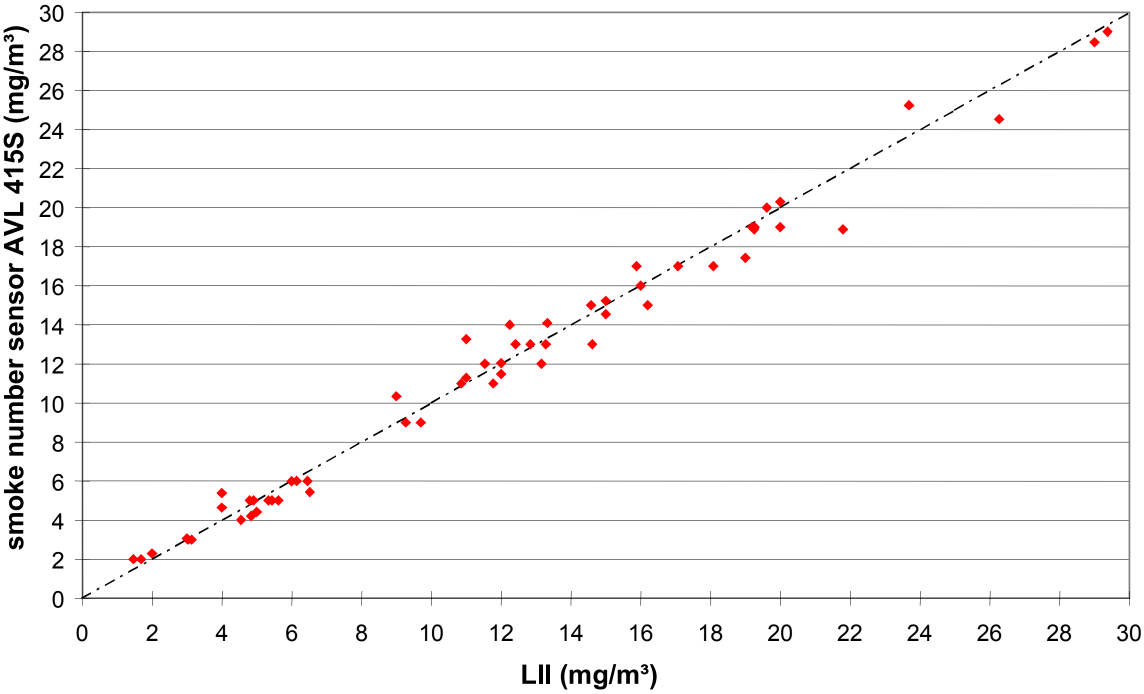

Fig. 5:. Comparison between the results of the LI²SA system and a smoke meter for a modern D.I. Diesel engine at stationary operation

In validating the performance of the LI²SA system for ultra-low concentrations, problems arise as standard measurement systems for particulate mass are currently not equipped for measurements in a range well below 0.1 mg/m³. Yet, investigations are underway to provide both calibration sources and reference experiments also for this concentration range. So far, an interesting number for characterising the LI²SA performance is the limit of detection (triple standard deviation of background signal), which currently is about 3µg/m³, but which can be even lowered - if desired - with a modified detection system.

Transient Tests

While the LI²SA sensor is also highly suitable for particulate measurements during stationary engine operation, special benefits can be obtained for instationary, highly-transient tests due to the high temporal resolution of the system.

An example is the application of the system to a truck engine (MAN D0826) on a test bench operated according to the European Transient Cycle (ETC). The LI²SA sensor was applied with a sampling rate of 4 Hz in various repetitions of this cycle. Part of the city-portion (total duration 10 minutes) for two independent runs is shown in Fig. 6, which demonstrates the excellent temporal response, dynamic range, and reproducibility of the system even under these highly transient conditions, which of course also requires a very good performance of the engine and the test bench.

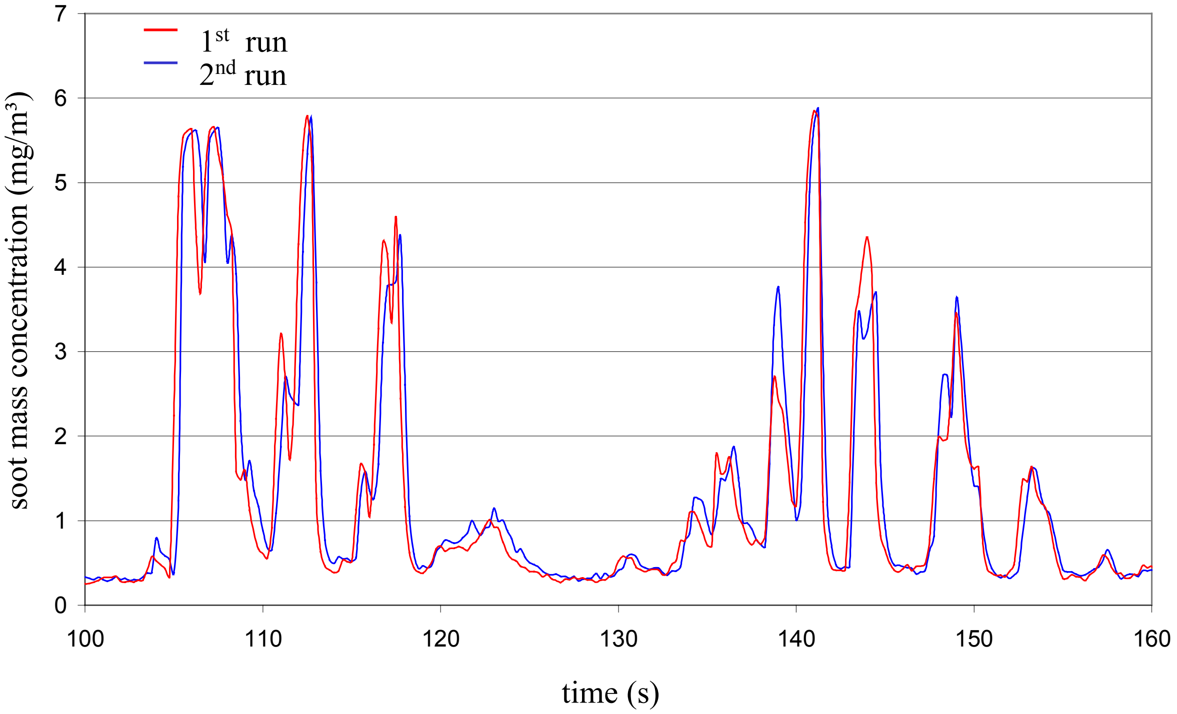

Fig. 6:. Performance of the LI²SA system during ETC test cycles (detail of the city portion)

Application with Chassis Dynamometers

Similar benefits of the LI²SA system may be stated for the application with chassis dynamometers, which form the standard platform for emission control of cars and trucks in usual customer-operation.

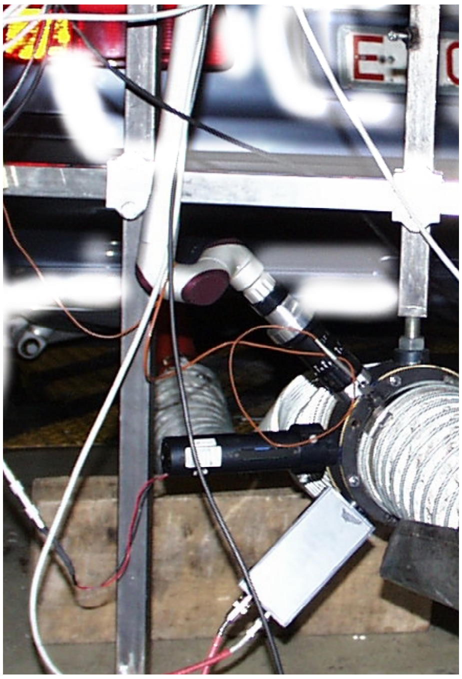

Fig. 7:. LI²SA on duty at a chassis dynamometer

Again, the LI²SA system may be very easily applied to a large variety of standard test cases (cf. Fig. 7). An example for its usefulness for the practical assessment of vehicle exhausts is shown in Fig. 8, where the effects of a provoked engine management malfunction can clearly be seen for a modern DI Diesel passenger car.

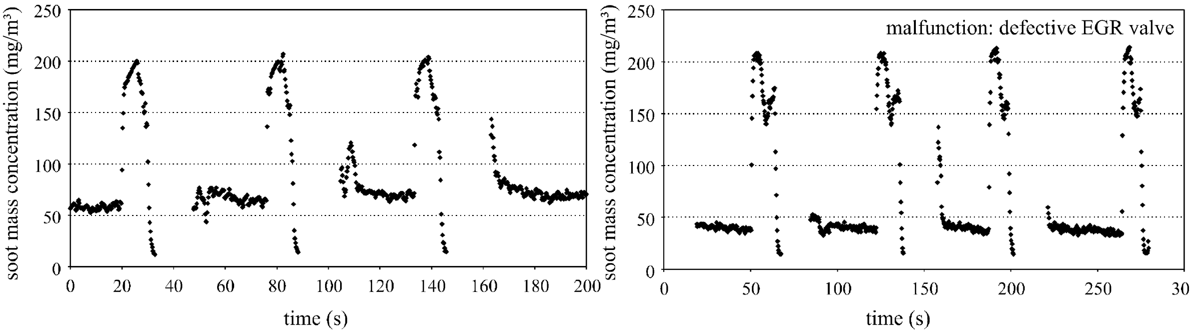

Fig. 8: Detection of enhanced soot emission for a passenger car due to a provoked malfunction of the EGR valve.

Conclusion

In order to meet the challenging requirements for the characterisation of particulates from modern low-emission engines, new measurement systems have to be applied. We have demonstrated that laser-induced incandescence may form a very suitable basis for advanced particulate measurement, as it allows a sensitive in situ determination of the most relevant carbonaceous particulate fraction.

Based on this measurement principle the LI²SA sensor system provides a flexible and easy-to-use means for engine and after-treatment development and the characterisation of engine and vehicle performance on test benches and chassis dynamometers, offering a data rate of several Hz for the real-time measurement also of highly transient processes. Besides measurement of particle mass LI²SA also yields information on specific particle surface, one important parameter for the assessment of possible health risks of emitted soot.

Acknowledgements

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References

1. D.B. Kittelson, J. Aerosol Sci. 29:575-588 (1998)
2. H. Burtscher, "Literature Study on Tailpipe Emission Measurements for Diesel Engines", 2001
3. K. Donaldson, X.Y. Li, and W. MacNee, J. Aerosol Sci. 29:553-560 (1998)
4. D. Kittelson and I. Abdul-Khalek, 2nd ETH Workshop on Nanoparticle Measurement, Zurich, 1998
5. L.A. Melton, Appl. Opt. 23:2201-2208 (1984).
6. S. Will, S. Schraml, and A. Leipertz, Twenty-Sixth Symposium (International) on Combustion (The Combustion Institute, Pittsburgh, 1996), pp. 2277-2284.

7. S. Will, S. Schraml, K. Bader, and A. Leipertz, Appl. Opt. 37:5647-5658 (1998).

8. S. Schraml, S. Will, and A. Leipertz, SAE Paper 1999-01-0146 (Society of Automotive Engineers, Warrendale, Pa., 1999).

9. S. Schraml, S. Will, A. Leipertz, T. Zens, and N. D.'Alfonso, SAE Paper 2000-01-2002 (Society of Automotive Engineers, Warrendale, Pa., 2000).

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Relation to possible Requirements, Benefits, Instrumentation

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5. ETH Workshop, Zürich 2001

Background

Broad basis of international activities on the application of LII to automotive exhaust gases

- National Research Programe **ABEME**
  (German Contribution to GRPE PMP activities)

  (Partners from Automotive Industry, Manufacturers of Measurement Systems, RW-TÜV, Research Institutes from toxicology/aerosol research and measurement)

  Financial Support by German Ministry of Education and Research (BMBF)
  German Engineering Federation (VDMA)

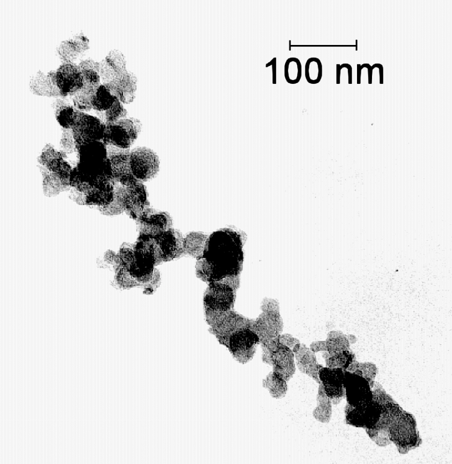
- Similar Activities by National Research Council of Canada (NRC)

- Broad theoretical framework by several workgroups worldwide

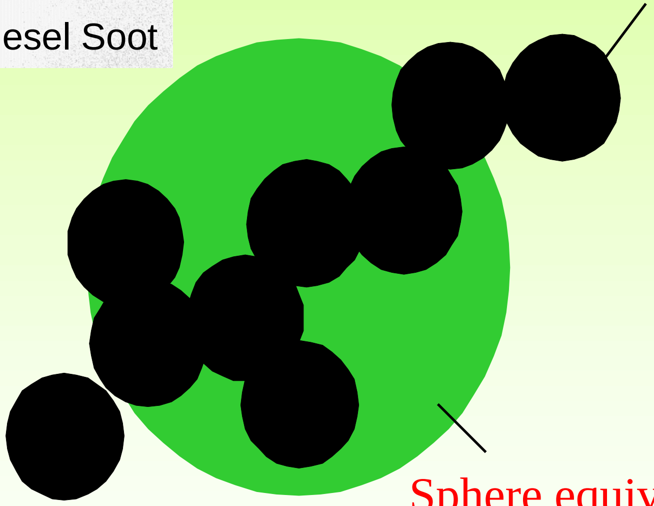
(Favorable) Possibilities of future PM measurement techniques

- EC as the most relevant component
  (adaptation of emission to air quality standards)
- directly mass related information and additional,
  effect related quantity (e.g., particle size, particle number,
  particle surface, ...)
- high sensitivity, applicable for ultra-low-emission engines
  within all relevant test cycles
- high temporal resolution, investigation of highly transient behaviour,
  better about 1 Hz
- highly specific on elemental carbon (EC), no interference with
  other exhaust components
- no influence of measurement conditions (temperature, dilution ratio),
  no condensation artifacts
- in-situ, online data evaluation (applicable also as a development tool)
- easy adaptation to different engines, no dilution tunnel necessary
- easy handling, reasonable price

Size Parameters



TEM-Image of Diesel Soot



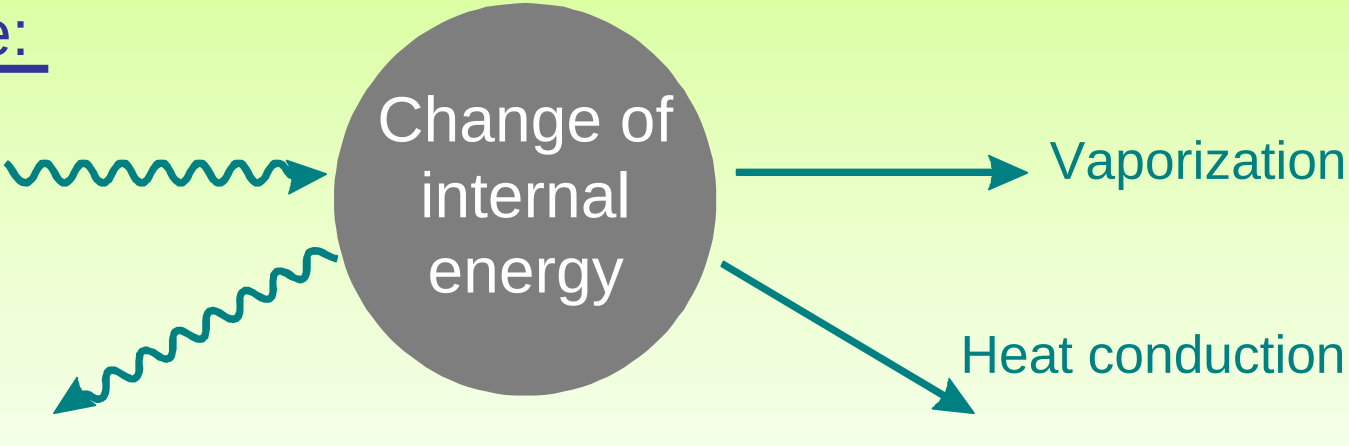
Specific Surface/
Primary Particle Size
exact definition, likely related to
chemical (surface) activity, transport
processes, remains unchanged after combustion

Sphere equivalent diameter (Aggregate/Agglomerate Size)
dependent on point of view
(e.g. diffusion diameter, aerodynamic diameter,
light scattering diameter, ..)
not constant during particle life time (coagulation)!
(→exact assessment of measurement conditions necessary)

Principle of the LII technique

Particle heating by a pulsed laser → **Detection of enhanced thermal radiation**

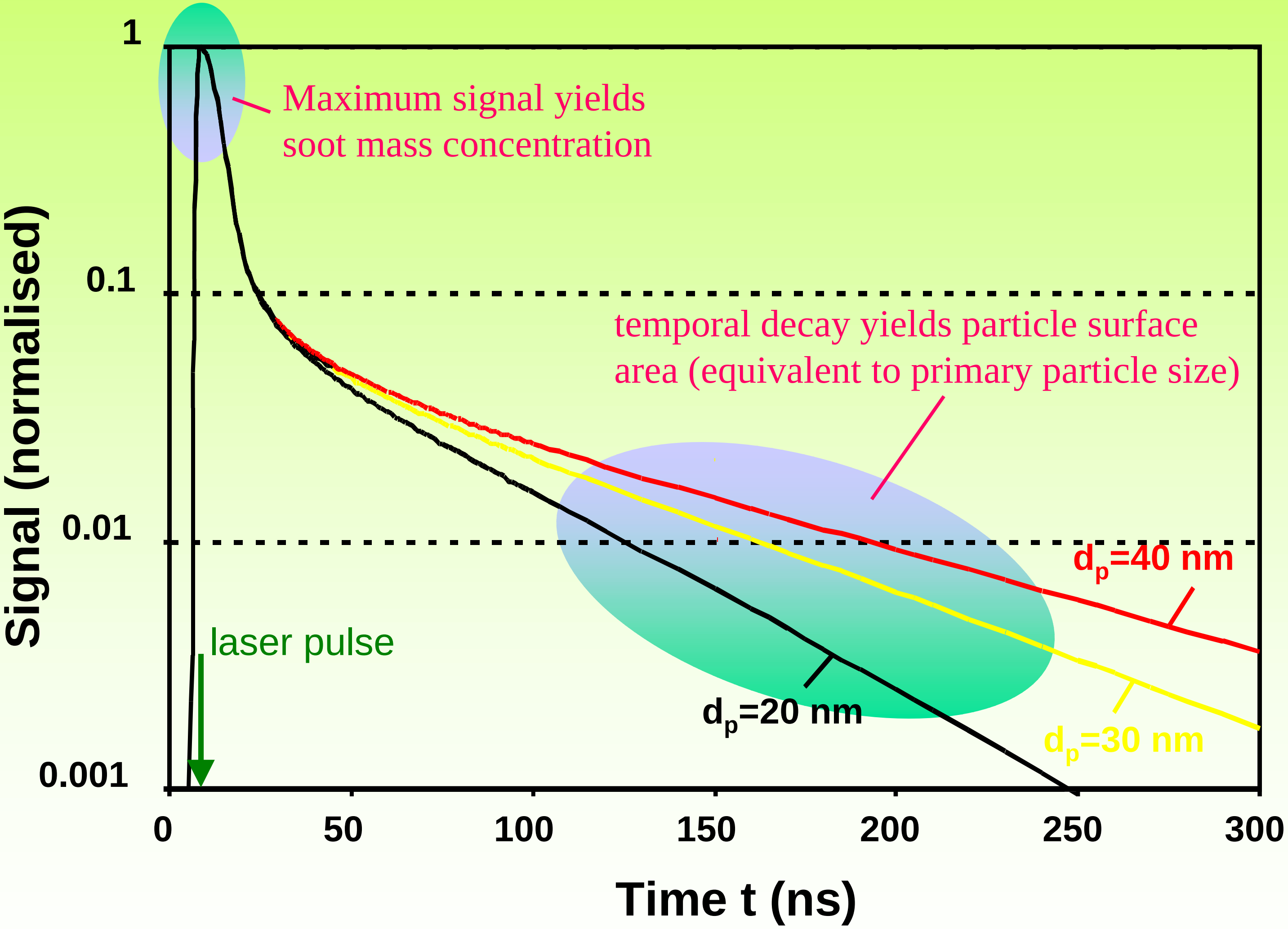
Power balance:



$$Q_{abs}\cdot\frac{\pi d_p{}^2}{4}\cdot E_i-\Lambda\cdot(T-T_0)\cdot\pi d_p{}^2+\frac{\Delta H_v}{M}\cdot\frac{dm}{dt}-\pi d_p{}^2\ \varepsilon(d_p,\lambda)\,M_\lambda{}^b(T,\lambda)d\lambda-\frac{\pi d_p{}^3}{6}\rho C\frac{dT}{dt}=0.$$

Absorption | Heat conduction | Vaporization | Thermal radiation | Change of internal energy

Measurement quantities of LII



Maximum signal yields soot mass concentration

temporal decay yields particle surface area (equivalent to primary particle size)

Measurement quantities of LII

Mass Concentration

- Maximum signal as a direct measure for soot mass concentration
- Only pure EC fraction is contributing (highly selective)
- independent of temperature, dilution ratio, ... (no conditioning, sampling, dilution necessary)
- Single calibration required

Surface Area/ Primary Particle Size

- Evaluation of signal decay Specific Surface Area/ Primary Particle Size
- no calibration necessary (only relative signal changes relevant)
- Comparison Experiment ⇔ Model
- Inclusion of exhaust temperature for signal evaluation

Other Quantities

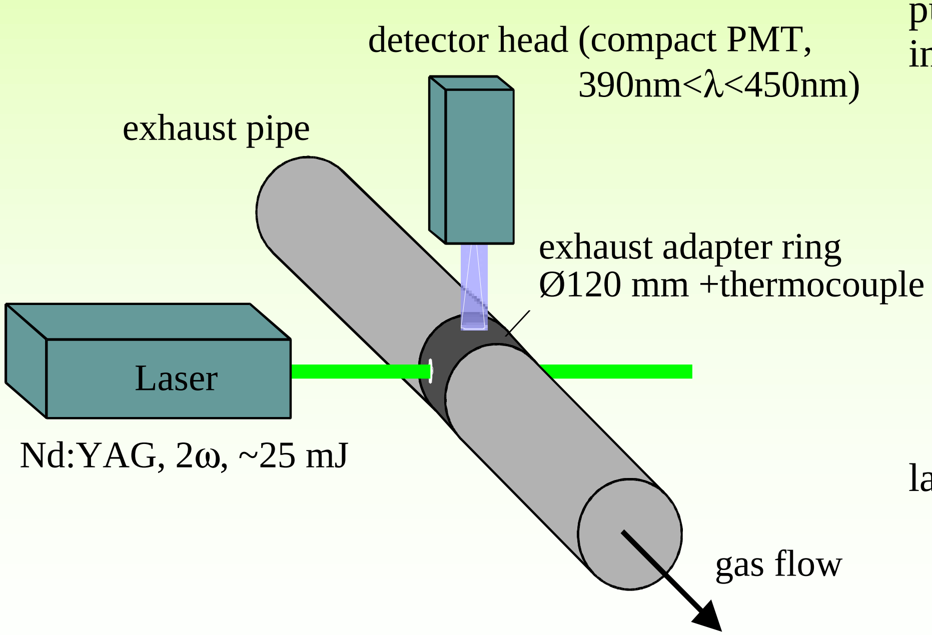
- Combination with elastic scattering: Agglomerate Size (optical and/or diffusion diameter)
- Totally emitted surface, number (densities) of primary particles and agglomerates

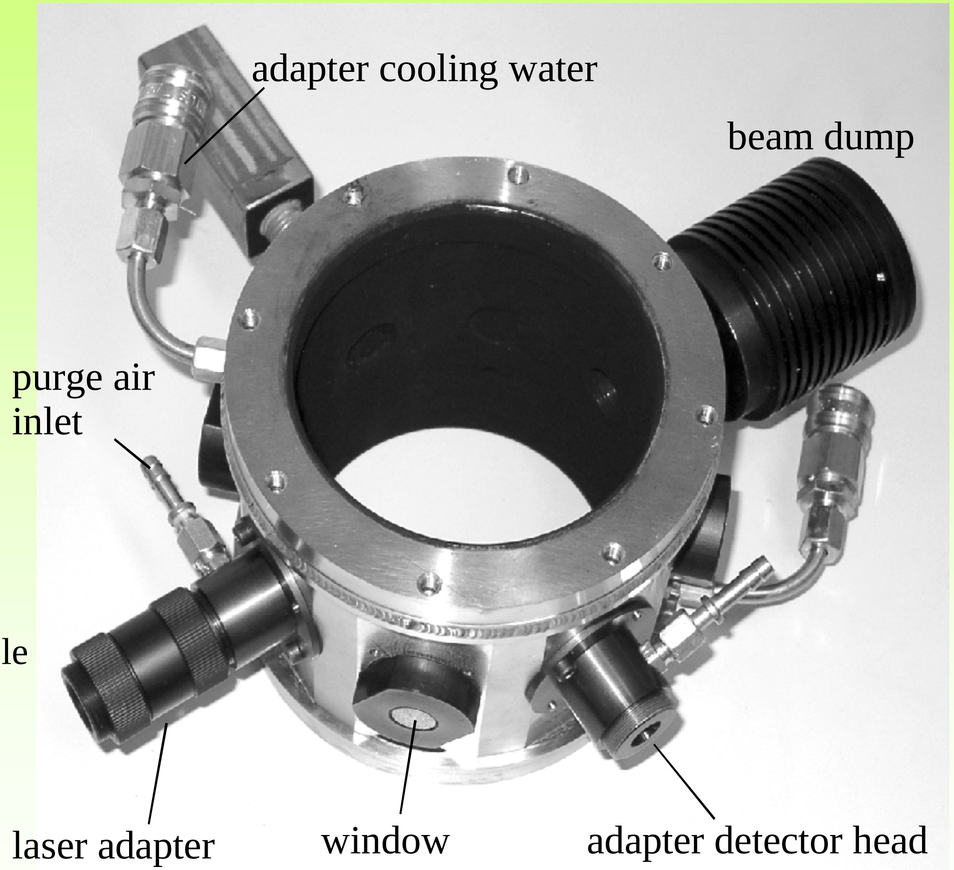
LII allows an easy and comprehensive characterization of EC particles

Sensor Concept

Direct integration of a adapter ring into the exhaust pipe

- no sampling procedure necessary
  (free of sampling artifacts)
- high temporal resolution
- integration of all sensor components
  (no further adjustment, easy handling)





Measurement Examples: Passenger Cars

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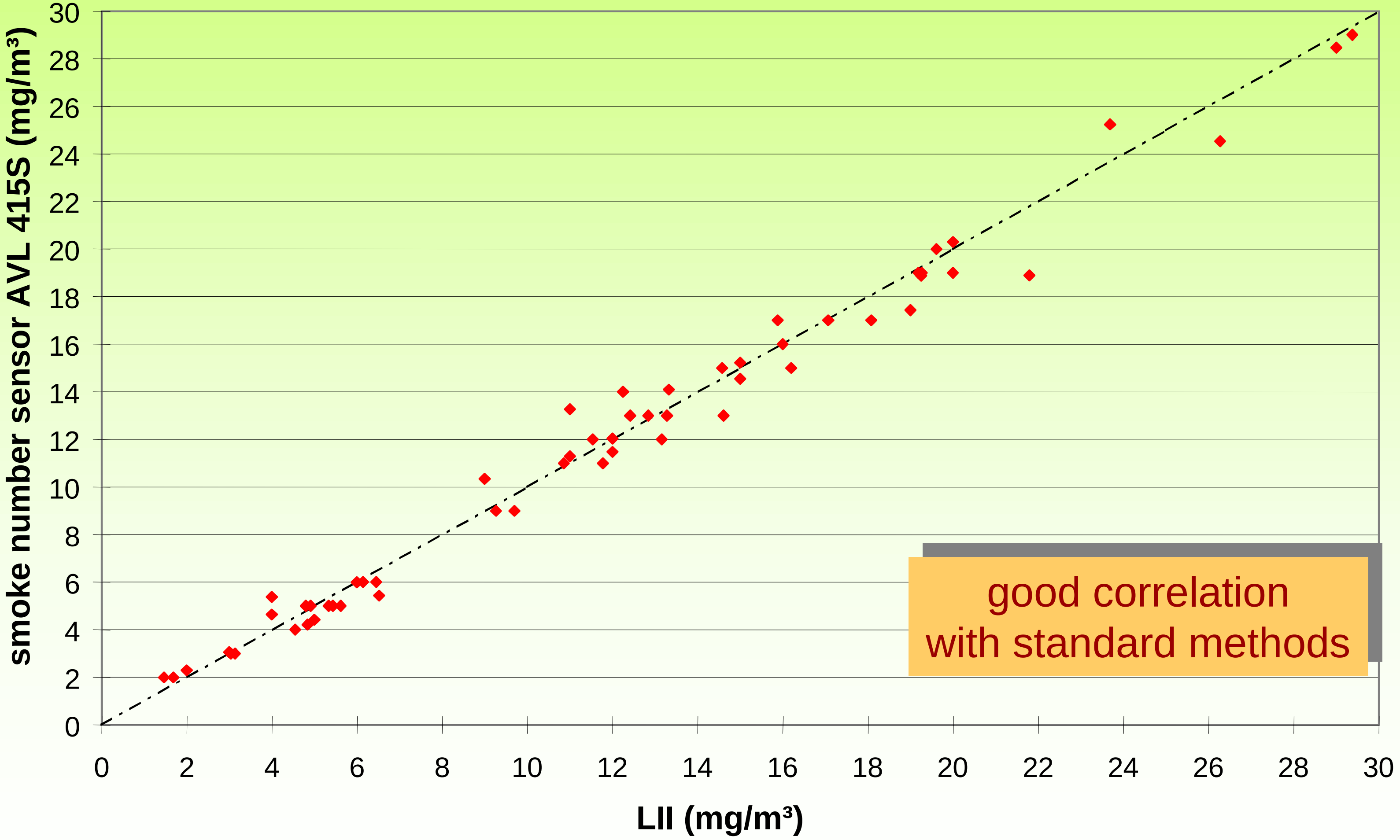
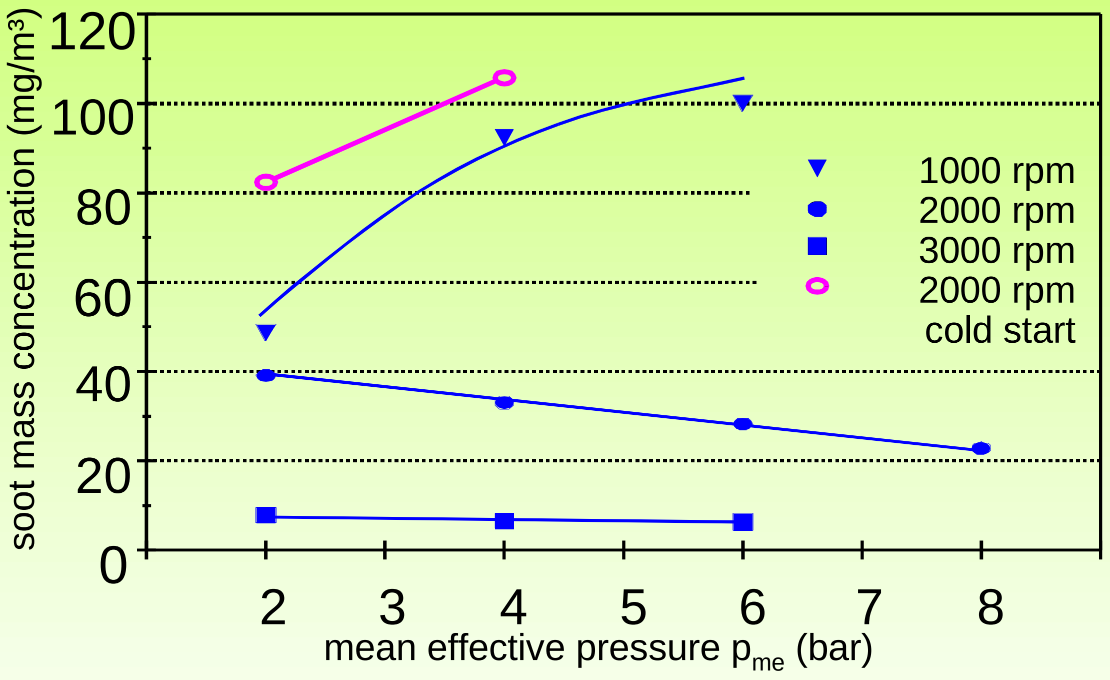
modern direct injection diesel engine (BMW 2,0l) on engine test bench



good correlation
with standard methods

Measurement Examples : Passenger Cars

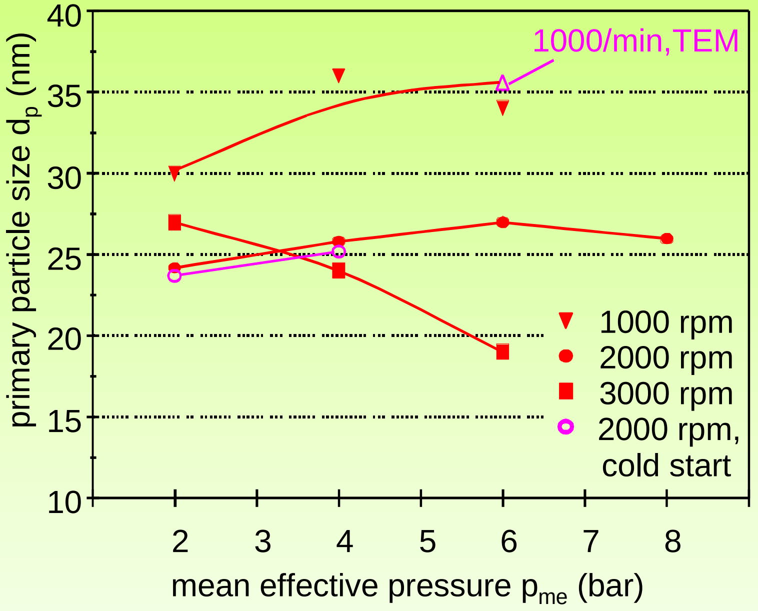
modern direct injection diesel engine (VW 1,9l TDI) on engine test bench

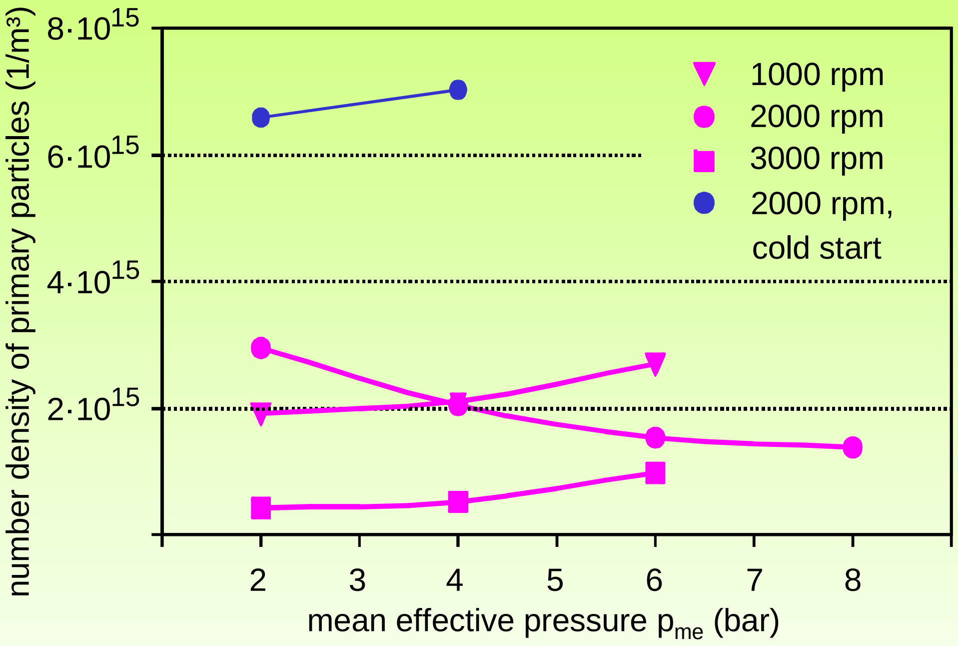


- increasing mass concentration for cold start conditions
- not clear: caused by larger particles ?
  or caused by increased number of particles?

Measurement Examples : Passenger Cars

simultaneous evaluation of signal decay yields additional information

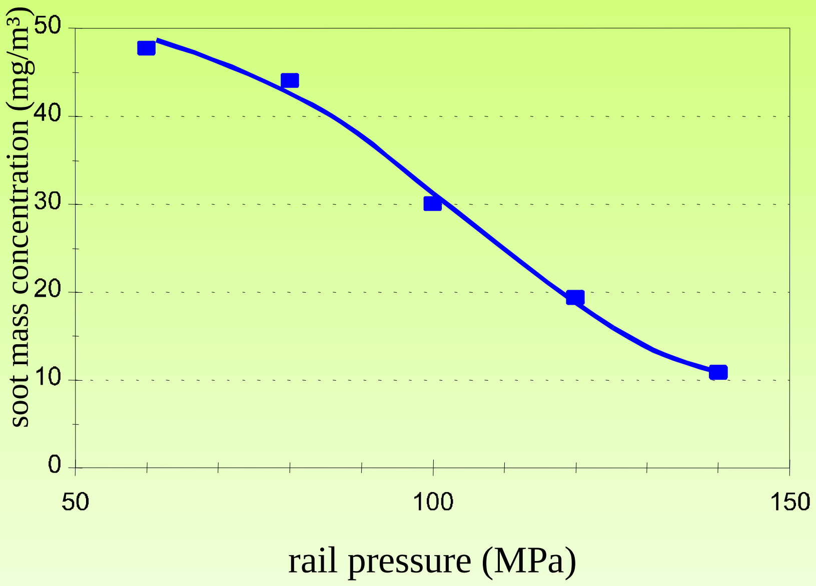


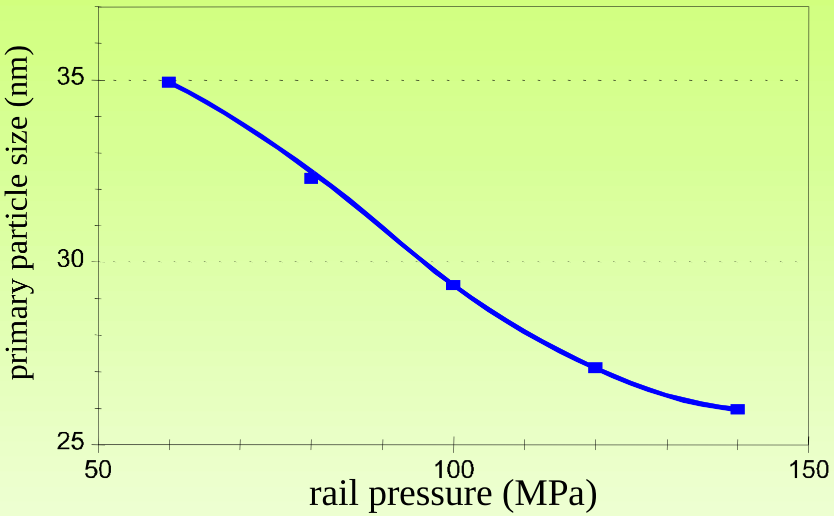


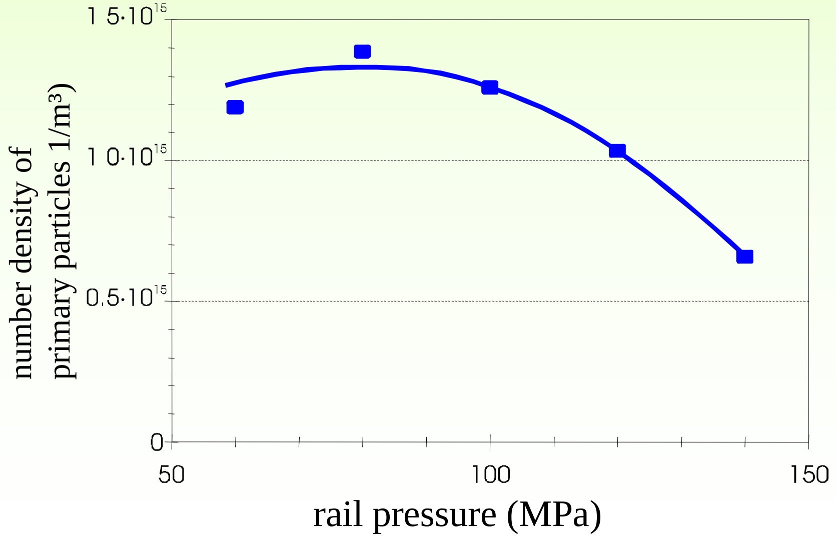
- increasing engine speed causes smaller primary particles/ larger surface (general trend)
- load dependence non-uniform (variable EGR-rate, SOI, ...)
- primary particle size / specific surface unchanged for cold-start condition
- cold start: increased number of primary particles
- good agreement with TEM results

Measurement Examples : Truck Engines

Influence of injection pressure on soot particles



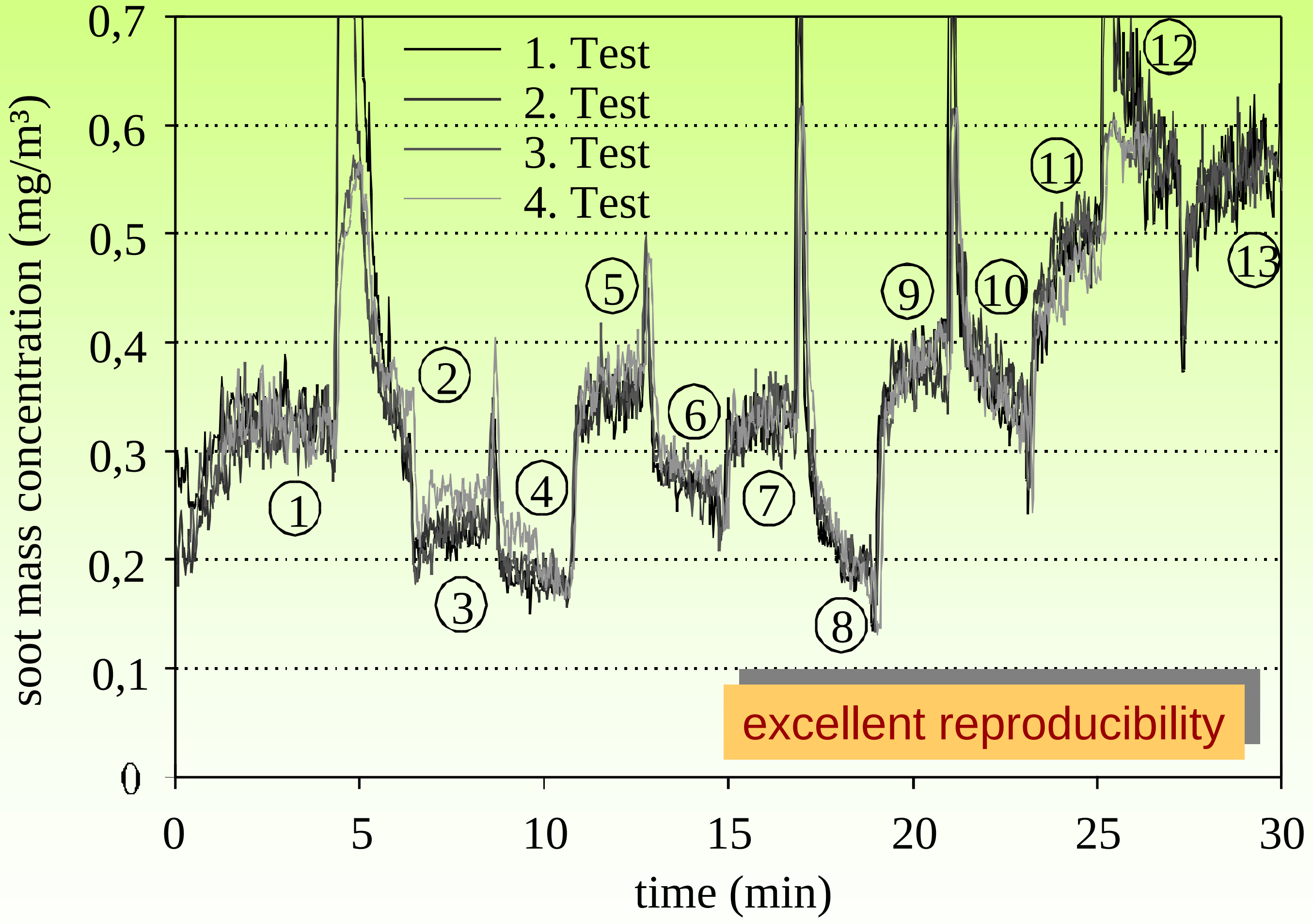




- increasing rail pressure causes reduction of soot mass concentration and larger specific surface/ smaller primary particle size
- but: number density of primary particles and totally emitted EC surface is also decreasing (opposite to previous studies with influence of artifacts from condensate)

Measurement Examples : Truck Engines

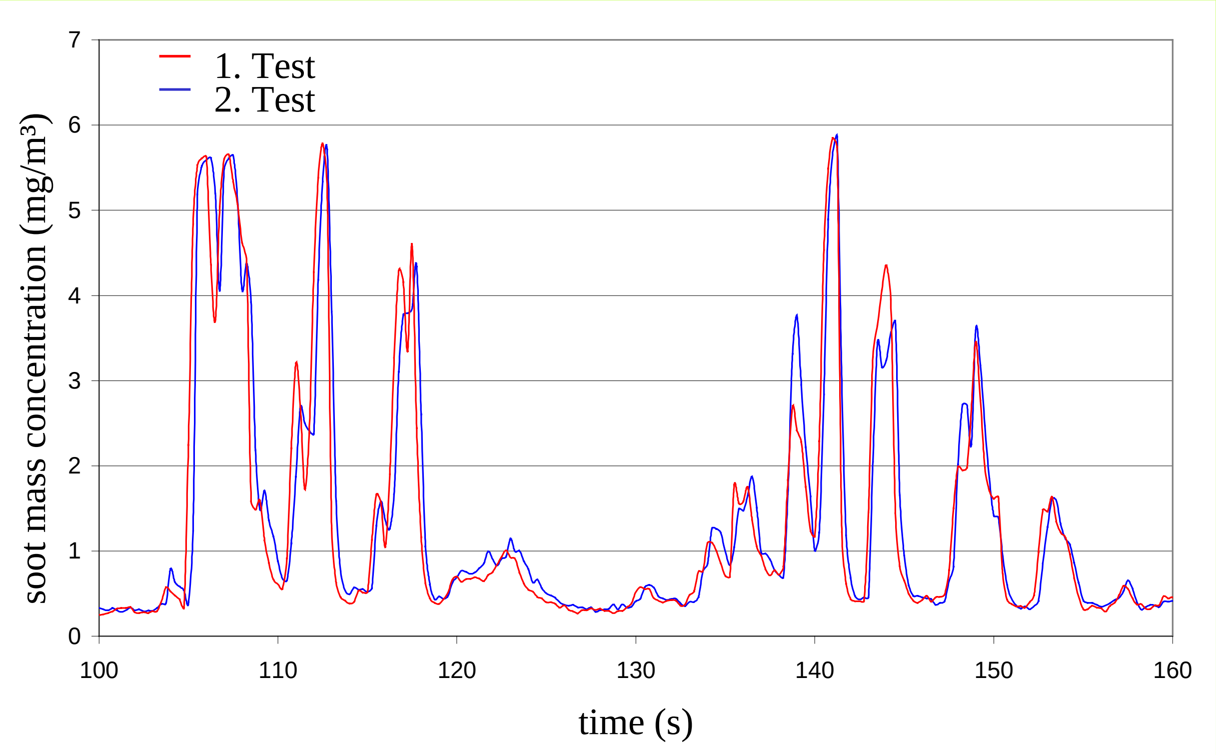
Mass concentration during multiple ESC test cycles



excellent reproducibility

Measurement Examples : Truck Engines

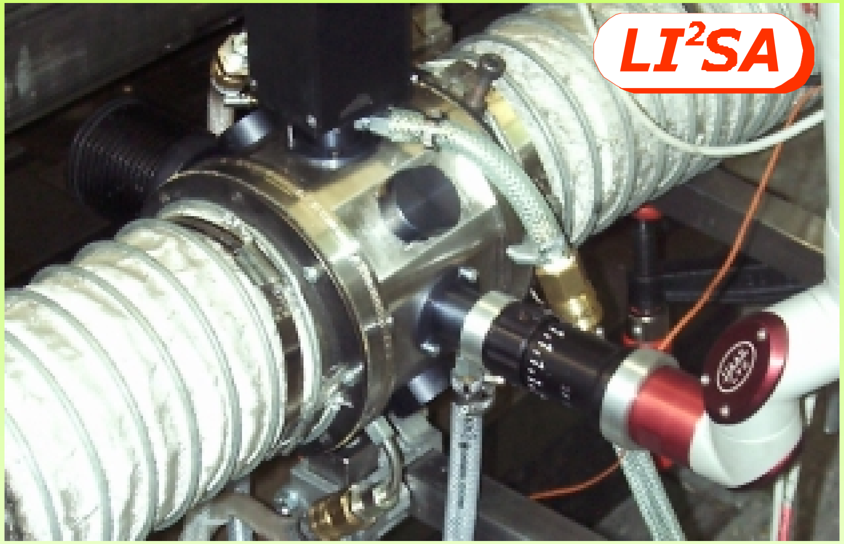
Mass concentration during multiple ETC test cycles (detail of city portion)



- good reproducibility even under highly transient conditions
- high temporal resolution (here: 4 Hz, nearly unlimited, given by laser repetition rate) (in-situ measurement without sampling)
- high dynamic range

Sensor Development / Cooperations

ESYTEC LTT ERLANGEN



ESYTEC Energie- und Systemtechnik GmbH, Erlangen

Conclusions, Sensor Features

Laser Induced Incandescence
Soot Analyzer LI²SA

- Simulateous measurement of EC mass concentration and specific surface area/primary particle size
- Highly specific: no influence from condensates or ashes
- Several extensions possible: agglomerate size (optical/diffusion diameter)
- Applicable for diluted <u>and</u> undiluted exhaust gases, no sampling artifacts
- High sensitivity: detection limit currently about 3µg/m³, 100µg/m³ already validated
- High temporal resolution: (currently) up to 20Hz
- Easy handling, direct application into the exhaust system, no further instrumentation necessary
- Robust setup, blinding free optical access,
- Fully automatic data acquisition and flexible on-line evaluation
- Application also as a development tool